



LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

Multi-wavelength investigation of laser-damage performance in KDP and DKDP following laser annealing

P. DeMange, C. W. Carr, R. A. Negres, H. B.
Radousky, S. G. Demos

September 7, 2004

Optics Letters

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

Multi-wavelength investigation of laser-damage performance in KDP and DKDP following laser annealing

P. DeMange*, C. W. Carr, R. A. Negres, H. B. Radousky*, and S. G. Demos

Lawrence Livermore National Laboratory, 7000 East Ave., Livermore, California 94551

**also with University of California, Davis Physics Department, 1 Shields Ave., Davis, California 95616*

(Received

The laser-induced damage performance of KDP and DKDP nonlinear crystals following pre-exposure to lower-energy laser pulses (laser annealing, also known as laser conditioning) is investigated as a function of wavelength for both, the damaging and conditioning pulses. To obtain a quantitative evaluation of damage performance of the material, we measure the density of damage events as a function of laser parameters. This new method allows for a detailed assessment of the improvement of material performance from laser conditioning and reveals the key parameters for optimizing performance depending on the operational wavelength.

Potassium dihydrogen phosphate (KH_2PO_4 or KDP) and its deuterated analog (DKDP) are the materials of choice for frequency conversion and electro-optical switching in large-aperture laser systems.^{1,2} Damage resistance testing of KDP and DKDP is currently used to evaluate the performance of individual crystal plates in large-aperture laser systems.³⁻⁵

The mechanisms of laser-induced damage and laser conditioning are still not well understood, although there is a growing indication that intrinsic and/or extrinsic defects play a key role.⁶⁻⁸ Laser-induced damage in KDP observed at laser fluences well below dielectric breakdown of the material manifests as sites localized to a few microns in diameter (henceforth referred to as pinpoints) which can reach temperatures as high as 1 eV during breakdown.⁹ This indicates highly localized absorption which may be attributed to either nanometer-sized particles of foreign material or intrinsic defect clusters. Experimental evidence has demonstrated the involvement of bulk defects prior to, during, and resulting from exposure to laser radiation.^{8, 10-13}

Laser annealing in KDP crystals using sub-damage threshold laser irradiation, commonly referred to as laser conditioning, has been reported to increase the material's resistance to damage at higher fluences.¹⁴ Laser conditioning is currently investigated as a method to increase the damage threshold of KDP and DKDP optics used in large-aperture laser systems,^{4,5} however, the laser parameters governing this effect that can be used to optimize this technique have not been previously prescribed.

In this work, we investigate the damage behavior of bulk KDP and DKDP under 1064 nm, 532 nm, and

355 nm irradiation subsequent to laser conditioning. The objective is to reveal the wavelength dependence and quantify the effectiveness of laser conditioning in order to achieve optimal performance for each operational condition.

The experiments were performed using the fundamental, second, and third harmonics of a pulsed Nd:YAG laser. The pulse durations (FWHM) of the output pulses were approximately 3.4 ns, 2.6 ns, and 2.5 ns for the 1064-nm, 532-nm, and 355-nm beams, respectively. The KDP and DKDP samples were cut to $1 \times 5 \times 5 \text{ cm}^3$ in size plates and polished on all sides. The samples tested were grown both conventionally and using the fast growth method. All results shown in this work were obtained from plates that they were cut from the same crystal and represent the typical behavior observed in all crystals.

The damage testing system used in this investigation has been detailed in a previous publication.¹⁵ The laser harmonics were aligned to co-propagate and focused by a 200-mm focal length cylindrical lens. The laser beams focused to a $1/e^2$ width of approximately 60 μm and height of 3000 μm , with less than ten percent standard deviation. An additional co-propagating 632.8-nm beam from a HeNe laser was focused by a 250-mm focal length cylindrical lens through the back of the crystals to over-fill the tested volume and illuminate any resulting damage pinpoints. Images of the scattered laser light were captured orthogonally to the direction of propagation of the lasers, through the side of the sample with a liquid nitrogen-cooled CCD camera using a long-working-distance microscope objective. With this arrangement, $\sim 22 \mu\text{m}^2$ of the sample illuminated area is projected onto each pixel of the CCD, allowing for detection of all damage pinpoints within the tested volume.

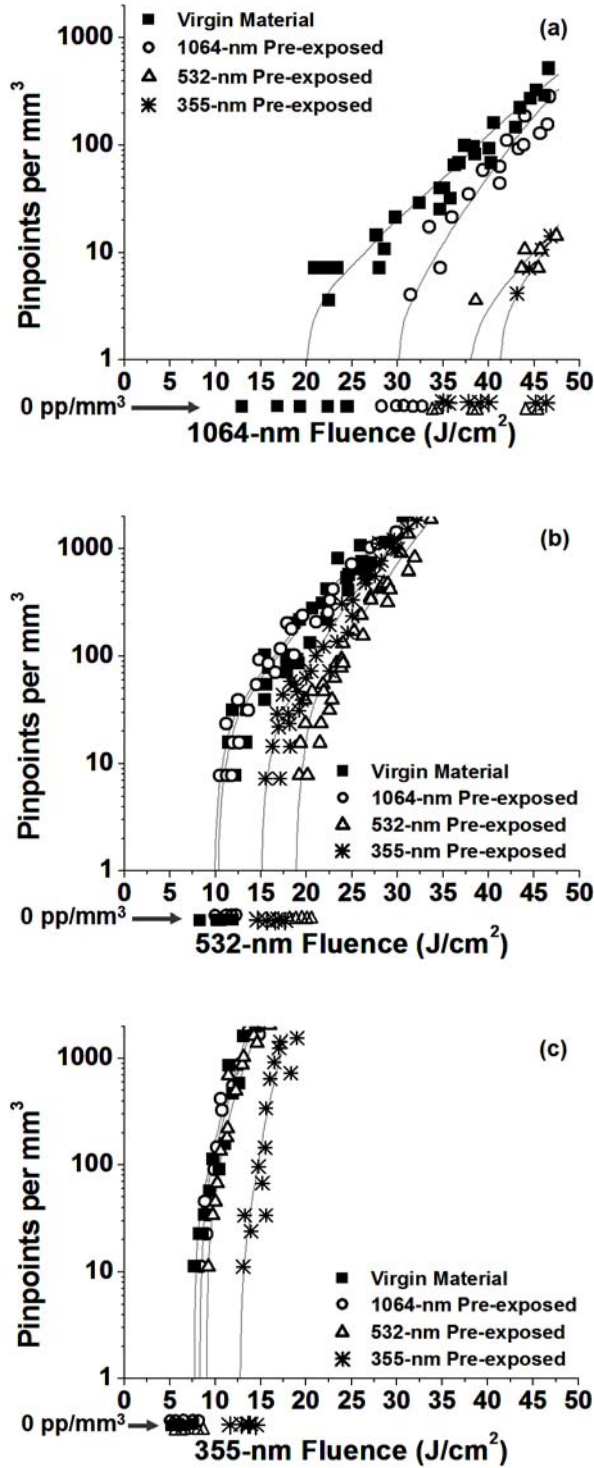


FIG. 1. Damage density profiles for a DKDP crystal sample on semi-log scale at 1064-nm (a), 532-nm (b), and 355-nm (c). The highest testing fluences for which no damage was observed (i.e., 0 pp/mm³) are indicated by the arrow.

Pristine sites were exposed to gradually increasing fluence levels in 1 J/cm² steps starting at 1 J/cm², with 10 pulses at each step. The fluence was ramped up to the highest fluence at which no damage resulted, as indicated by the CCD camera. This highest conditioning fluence was approximately 11 J/cm² at 355 nm, 17 J/cm² at 532 nm, and 24 J/cm² at 1064 nm. Due to energy fluctuations in each laser pulse and the statistical nature of ns-pulse damage initiation in KDP and DKDP, the highest average fluence reached during conditioning was generally around 2 J/cm² below the damage onset fluence at 532 nm and 355 nm and around 6 J/cm² below at 1064 nm. The conditioned sites were tested with one damaging pulse and an image of the damage was captured for analysis of the resulting damage pinpoint density. This same conditioning protocol was repeated at different sites in the bulk with the fluence of the damaging pulse varying at each site.

The damage pinpoint density was measured over a volume of $\sim 0.1 \text{ mm}^3$, representing the region of the crystal exposed only to peak laser fluence (within 5%, i.e. $\sim 338 \text{ }\mu\text{m}$ transverse width). The resulting pinpoint density was then plotted versus the peak damaging fluence. The lowest testing fluence at which damage was observed we will refer to as the damage onset fluence (DOF).

Figure 1 shows the profiles of damage pinpoint density as a function of fluence for damage testing at 1064-nm, 532-nm, and 355-nm irradiation. These profiles demonstrate the damage behavior of a DKDP sample both with conditioning and without conditioning using the fluence-ramping method. The plots are on a semi-log scale and data fit lines are drawn in as a guide to the eye.

The pinpoint density profiles obtained under 1064-nm damage testing are shown in Fig. 1(a). The DOF of the unconditioned material is found to be $\sim 20 \text{ J/cm}^2$. Pre-exposure at 1064 nm demonstrates a substantial increase in the DOF to 30 J/cm². Pre-exposure at 532 nm increases the DOF even more, reaching 37.5 J/cm², and 355 nm pre-exposure shows at least a factor of two increase. The pinpoint density profiles obtained under 532-nm damage testing are shown in Fig. 1(b). The DOF of the unconditioned material is found to be $\sim 10 \text{ J/cm}^2$. An increase in the DOF to 15 J/cm² is achieved by pre-exposure at 355 nm while pre-exposure at 532 nm shows a larger increase in the damage threshold to nearly 19 J/cm². The pinpoint density profiles obtained under 355-nm damage testing are shown in Fig. 1(c). These results show that pre-exposure at 355 nm increased the DOF to above 12.5 J/cm² from the $\sim 8 \text{ J/cm}^2$ measured in the unconditioned material. However, pre-exposure at 1064 nm and 532 nm results in no noticeable increase in the 355-nm DOF. All pinpoint density curves are

shifting to higher fluence with conditioning representing the change in the damage initiation threshold to the entire population of defect responsible for damage initiation at different fluence.

Our experimental data illustrate the conditions needed to achieve optimal level of improvement to the damage performance in bulk KDP and DKDP. As a general rule, conditioning occurs for pre-exposure at photon energies equal to or higher to that used for damage testing. Fig. 1(b) shows that pre-exposure to 532-nm pulses provides better conditioning for 532-nm operation than pre-exposure to 355-nm pulses, an opposite trend to that observed in Fig. 1(a), where the lower wavelengths provided better conditioning for 1064-nm testing. Damage testing at 355 nm indicated that pre-exposure at either 1064 nm or 532 nm provides no improvement to damage resistance while, in contrast, pre-exposure at 355 nm shows significant improvement.

Recent results have highlighted the potential role of either clusters of different intrinsic defects and/or various species of impurities in laser-induced damage initiation.^{6,7,16} From our results, it is difficult to reason that there is only one species of defects in KDP, intrinsic or extrinsic, responsible for damage initiation at all wavelengths. For example, it is shown that laser light at 355 nm improved damage performance at 532 nm, but not the reverse. One may then assume that the species of defect responsible for damage initiation at 532 nm absorbs the 355-nm laser light resulting in conditioning. The species of defects responsible for damage initiation at 355 nm must then be different since they do not absorb the 532-nm laser light to result in conditioning. Similarly, we demonstrated that laser light at 355 nm and 532 nm improve damage performance at 1064 nm, but not the reverse in either case. As seen in Fig. 1 for all three cases, one wavelength improves damage performance at another wavelength but not vice-versa. In addition, it has been reported that thermal annealing improves damage performance for operation at 1064 nm but not for 355 nm.¹⁷ The above experimental evidence may indicate that there are in fact different species of defects responsible for damage initiation in bulk KDP and DKDP at different wavelengths.

In conclusion, a quantitative investigation of the improvement of damage performance of bulk KDP and DKDP under pre-exposure to laser irradiation reveals the level of conditioning versus pre-exposure laser fluence and wavelength. Using the fluence-ramping conditioning technique, the damage onset fluence is observed to increase by at least 50-100%, for each wavelength tested. Pre-exposure using fluence ramping to above the damage onset fluence may show an even larger improvement to the damage performance, as well as provide conditioning for the

wavelengths tested in which no conditioning was observed. However, the efforts here have focused toward improving the damage performance without damaging the material. These results demonstrate the level of improvement that may be achieved at different wavelength operation and that the effectiveness of conditioning is strongly dependent on the conditioning wavelength. This work reveals the conditioning behavior of these important materials but it does not address the mechanisms for laser conditioning. This will be the focus of future work.

This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under contract W-7405-Eng-48.

- ¹ N. P. Zaitseva, J. J. De Yoreo, M. R. Dehaven, R. L. Vital, K. E. Montgomery, M. Richardson, and L. J. Atherton, *J. of Cryst. Growth* **180**, 255 (1997).
- ² J. J. De Yoreo, A. K. Burnham, P. K. Whitman, *Int. Mater. Rev.* **47**, 113 (2002).
- ³ A. K. Burnham, M. Runkel, M. D. Feit, A.M. Rubenchik, R. L. Floyd, T. A. Land, W. J. Siekhaus, and R. A. Hawley-Fedder, *Appl. Opt.* **42**, 5483 (2003).
- ⁴ L. M. Sheehan, M. R. Kozlowski, F. Rainer, and M. C. Staggs, *Proc. SPIE* **2114**, 559 (1994).
- ⁵ F. Rainer, F. P. DeMarco, M. C. Staggs, M. R. Kozlowski, L. J. Atherton, and L. M. Sheehan, *Proc. SPIE* **2114**, 9 (1994).
- ⁶ C. W. Carr, H. B. Radousky, S. G. Demos, *Phys. Rev. Lett.* **91**, 127402 (2003).
- ⁷ C. S. Liu, N. Kioussis, S. G. Demos, and H. B. Radousky, *Phys. Rev. Lett.* **91**, 015505 (2003).
- ⁸ S. G. Demos, M. Staggs, and H. B. Radousky, *Phys. Rev. B* **67**, 224102 (2003).
- ⁹ C. W. Carr, H. B. Radousky, A.M. Rubenchik, M.D. Feit, and S. G. Demos, *Phys. Rev. Lett.* **92**, 087401 (2004).
- ¹⁰ S. G. Demos, M. Yan, M. Staggs, J. J. De Yoreo, H. B. Radousky, *Appl. Phys. Lett.* **72**, 2367 (1998).
- ¹¹ J. E. Davis, R. S. Hughes Jr. and H. W. H. Lee., *Chem. Phys. Lett.* **207**, 540 (1993).
- ¹² C. D. Marshall, S. A. Payne, M. A. Henesian, J. A. Speth, and H. T. Powell, *JOSA B* **11**, 774 (1994).
- ¹³ M. M. Chirila, N.Y. Garces, L.E. Halliburton, S.G. Demos, T.A. Land, H.B. Radousky, *J. Appl. Phys.* **94**, 6456 (2003).
- ¹⁴ J. Swain, S. Stokowski, D. Milam, and F. Rainer, *Appl. Phys. Lett.* **40**, 350 (1982).
- ¹⁵ P. DeMange, C. W. Carr, H. B. Radousky, and S. G. Demos, *Rev. Sci. Instr.*, in press.
- ¹⁶ M.D. Feit and A.M. Rubenchik, *Proc. SPIE* **5273**, 74 (2003).

- ¹⁷ M. Runkel, S. Maricle, R. Torres, J. Auerbach, R. Floyd, R. Hawley-Fedder, and A. K. Burnham, Proc. SPIE **4347**, 389 (2001).